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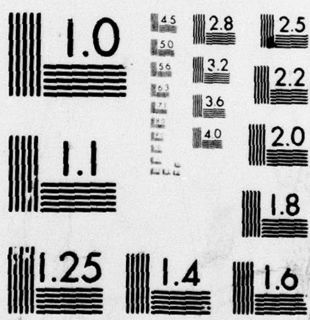

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Work is reported in the areas of:
(1) Collisional studies using laser spectroscopy;
(2) The nature and validity of collision kernels; → next page

(3) Strong field effects in laser spectroscopy,
(4) Collisionally-aided radiative excitation including a model potential calculation,
(5) Radiative collisions including model potential calculations,
(6) Collisional studies using photon echoes or Optical Ramsey Fringes,
(7) Photon echoes produced by standing-wave fields, and
(8) Photo-ionization by pulsed laser fields.

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Research has been carried out in the areas of (1) Laser Spectroscopic Collisional Studies, (2) Collisionally-Aided Radiative Excitation, (3) Radiative Collisions, (4) Photon Echoes in Standing-Wave Fields, and (5) Photoionization in Pulsed Fields. Our results are summarized briefly below; details of the calculations are contained in the reprints or preprints appended to this report.

1. Laser Spectroscopic Collision Studies (P. Berman)

A detailed analysis of the laser spectroscopy of 3-level systems has been undertaken. The problem considered is that in which a laser field (pump) of arbitrary strength excites a given transition in some "active" atoms while a second laser field (probe) acts on a transition coupled to the first. The active atoms are immersed in a perturber bath and undergo collisions with the perturbers. The nearly monochromatic pump field excites a narrow longitudinal active-atom velocity distribution, and one can monitor the collisional relaxation of this distribution by studying the probe absorption profile. The purpose of our analysis is to determine the usefulness of laser spectroscopic methods in determining total scattering cross sections, differential scattering cross sections, and interatomic potential curves.

Theoretically, we have derived[1*] saturation spectroscopy line shapes valid in the strong pump field - weak probe field limit, assuming that collisions are phase-interrupting in their effect on level coherences and velocity-changing in their effect on level population densities. The velocity-changing collisions are treated in a large-angle scattering limit and specific calculations are carried our for the Keilson-Storer collision kernel.[1]

Among the new results obtained are:

i) In strong pump fields, collisions can result in increased peak and

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* Asteriks on references indicate reprints or preprints appended to this report.

total probe absorption. This result has implications for increasing yields in certain laser isotope separation schemes.

ii) Velocity-changing collisions can significantly distort or eliminate the ac Stark splitting that occurs in laser spectroscopic profiles.

iii) Laser spectroscopic profiles are sensitive enough to provide limited information on the interatomic potentials giving rise to the large-angle-scattering.

iv) The line profiles can be used to test the validity of various macroscopic collision kernels and rates that are used to parametrize atomic and molecular systems.

Experimentally, in collaboration with Drs. P. Liao and J. Bjorkholm at Bell Telephone Laboratories, we have carried out a systematic study of the laser excitation of a "three-level" system ($3S_{1/2} - 3P_{1/2} \rightarrow 4D_{3/2}$) in Na atoms perturbed by various rare gas perturbers.[2*] This work represents the first systematic study of velocity-changing collisions in short-lived excited states using laser saturation spectroscopy.

The results of this research may be summarized as follows:

i) By using pump detunings (difference between pump laser and atomic transition frequencies) greater than the Doppler width associated with the transitions, one can determine various pressure broadening and shift coefficients which may be related to total scattering cross sections. In addition one can observe the process of collisionally-aided radiative excitation (see below) with such detunings.

ii) With detunings less than the Doppler width, various collision kernels can be tested.

iii) For Na-He collisions, both the Keilson-Storer and hard-sphere collision kernels adequately describe large-angle-scattering. For Na-Ne and Na-Kr collisions, the hard-sphere kernel is superior to the Keilson-

Storer one; however neither kernel leads to good agreement over the entire line profile. These results imply that there is a strong, attractive part to the short-range Na-Ne and Na-Kr potential curves. Additional evidence for an attractive well in the Na-Ne system has recently been discovered.[3]

iv) Fine-structure state-changing collisions in Na are important and must be incorporated into the theory.

v) Optical pumping effects are also important.

vi) Cross sections for velocity-changing collisions between Na ($3P_{1/2}$) and He, Ne and Kr perturbers were obtained.

The major result of this work is that it demonstrates that saturation spectroscopy can be used as a quantitative probe of atomic collisions.

2. Collisionally-Aided Radiative Excitation (S. Yeh, E. Robinson, P. Berman)

The work on Collisionally-Aided Radiative Excitation (CARE) in two-level systems[4*] has been extended to three-level ones.[5*] In three-level CARE, two pulsed laser fields are incident on a three-level atom. The fields are off-resonant; there is no atomic excitation in the absence of collisions. Collisions can shift the energy levels into instantaneous resonance with the applied fields leading to an enhancement of the excitation cross sections.

In three-level systems, both "stepwise" (SW) and "two-quantum" (TQ) excitation processes are possible. The interference of these two excitation paths during CARE can lead to new effects. One such effect is an oscillation in the <u>total</u> CARE cross section as a function of interatomic velocity. This oscillation is a new feature of CARE, not previously predicted, and can be used to obtain information on interatomic potentials.[5] Another result obtained is an asymptotic dependence on detuning that is intermediate to those predicted for pure SW or TQ processes. The above work was carried out in the weak-field limit, and an extension to the strong-field case

is envisioned. In laser isotope separation schemes using off-resonant photons, CARE may provide a means for increasing yields.

A model-potential calculation for CARE in two-level systems in the weak field limit has also been performed.[6*] Since computer calculations of CARE are costly, it is extremely useful to have analytic results for interatomic potentials that approximate the true potentials reasonable well. Analytic solutions have been obtained using either sech or $(\mathrm{sech})^2$ type potentials. These potentials are smooth and can be parameterized to closely approximate van der Waals interaction potentials.

3. Radiative Collisions (E. Robinson)

In radiative collisions, laser photons are used to provide the energy difference to allow certain collisional processes to become energetically possible. The equations that describe radiative collisions are quite similar to those for CARE and can be solved by similar techniques. As in the case of CARE, computer solutions are costly and it is advantageous to have analytically solvable models that take into account both the collisional coupling <u>and</u> shift of the levels involved in the transition.

Two analytically solvable model potentials have been found that can be paremeterized to approximate known potentials. Both models take the coupling potential to be of the form of a hyperbolic secant. The level-shifting potential is taken to be a delta function in one model[7*] and a hyperbolic secant squared in the other one.[6] Calculations carried out in the weak field limit[6,7] are in good agreement with numerical calculations using van der Waals potentials and also in good agreement with experiment.[6] The models correctly predict the asymmetries observed in radiative collision profiles. Attempts to extend both models to the strong-field case are currently in progress.

4. Photon Echoes in Standing-Wave Fields (P. Berman)

In collaboration with J. LeGouet of Laboratoire Aime Cotten, we have calculated the response of an atomic system subjected to two nearly-resonant strong standing-wave fields separated in time.[8*] If the pulses are separated by a time T, one predicts echoes at times nT (n=integer) following the second pulse. These pulses are similar in nature, but not identical, to Optical Ramsey Fringes. A study of the amplitude of the echoes serves to monitor relaxation processes occurring in the gas. The echoes are sensitive to collisions that affect <u>both</u> level coherences and level population densities, in contrast to "normal" photon echoes which reflect collisional effects on level coherences only. The usefulness of standing-wave echoes for standards work is still to be explored.

A related result contained in our work, but not noted explicitly, is the appearance of echoes in <u>populations</u> as well as level coherences. These echoes, reflecting harmonics excited by the standing-wave fields, occur at various times following the second pulse. For the population modulated in space at one-half the excitation wave length, the echoes occur at times (n/2)T following the second pulse. These population echoes were predicted by Mossberg, Kachru, Wittaker and Hartmann.[9] Both the population and standing wave echoes have been recently found experimentally.[9]

5. Photoionization in Pulsed Fields (E. Robinson)

It has been discovered that it may be possible to observe "mock" structure in resonant multiphoton ionization using pulsed fields.[10*] For this process to occur, the field must be nearly resonant with a given transition. The field excites this transition and produces an oscillating probability amplitude in an excited state. If photoionization occurs from this intermediate state, the oscillatory pattern in the excited state amplitude may be mirrored in the photoionization cross section. The process is favored when the pulse area is approximately 2π and when several photons are required to produce photoionization from the excited state.

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